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Spontaneous Decay of an Atom Near a Phase Conjugator

by

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SPONTANEOUS DECAY OF AN ATOM NEAR A PHASE CONJUGATOR

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ABSTRACT

Relaxation of the density operator of an atom near a four-wave mixing phase conjugator (PC) is studied. The relaxation operator  $\Gamma$  can be expressed in the Cartesian components of the atomic dipole operator and correlation functions of the vacuum electromagnetic field. We evaluate these correlation functions in terms of the Fresnel coefficients for reflection and transmission of a polarized monochromatic plane wave. Our expression for  $\Gamma$  includes both linear and nonlinear interactions in the medium, and it reduces to well-known results in the limits of a pure dielectric, a mirror, and empty space. The example of a model two-state atom in combination with a model PC is worked out in detail. From the rate equations for the populations of the levels we infer that the relaxation acquires a contribution from both ordinary spontaneous decay and stimulated transitions which are induced by the PC. It is shown that an atom in its ground state has a finite probability of being excited, and that the excited state has a finite population in the long-time limit. We also work out the case of a degenerate two-level atom in combination with a realistic model of a transparent PC, and it is shown that our present results



## I. INTRODUCTION

An excited atom can decay spontaneously to a lower state, in combination with the emission of a fluorescent photon, due to the coupling of its dipole moment  $\underline{\mu}$  with the electromagnetic field of the vacuum. For a model two-state atom with excited state  $|e\rangle$  and ground state  $|g\rangle$ , and level separation  $\hbar\omega_0$ , this amounts to an effective lifetime  $1/A$  of the excited state, where

$$A = \frac{\omega_0^3}{3\pi\epsilon_0\hbar c^3} |\langle e|\underline{\mu}|g\rangle|^2, \quad (1.1)$$

equals the Einstein coefficient for spontaneous decay. This relaxation constant  $A$  pertains to an atom in empty space. When an atom is close to the surface of a dielectric or a mirror, with the atom-surface normal distance of the order of an optical wavelength (which is large compared to the atomic dimensions), then the expression for the lifetime of an excited state changes dramatically [1-12]. Such an alteration of lifetimes due to the presence of media was first demonstrated by Drexhage for optical transitions in dye molecules in thin films [13]. Much later, lifetime measurements were performed on a single Rydberg atom in a microwave cavity [14,15], and for an ordinary atom near a mirror or in between parallel mirrors [16,17]. It appeared, in agreement with theory, that both an enhancement and an inhibition of the spontaneous decay rate could be obtained.

In order to understand this phenomenon one merely has to realize that a photon, which is emitted by the atom into the direction of the surface, has a probability of being reflected and a probability of being refracted, just as in classical optics. A reflected wave then interferes with photons which are emitted in a direction away from the medium, and this can give rise to either

a reduction or an increase of the fluorescent intensity. Since energy conservation requires that the energy which is emitted by the atom as radiation must be provided by that atom, we conclude that a destructive (constructive) interference of radiation must be accompanied by an increase (decrease) of the lifetime of the atomic state. From a different point of view, we can say that the electromagnetic vacuum near a medium is different from the vacuum of empty space. Without the medium, the electric-field operator in the Heisenberg picture is a superposition of travelling plane waves, without any preference for a specific orientation in space. An excited atom can emit a fluorescent photon in any of the available plane-wave modes, and these photons travel away from the atom (their superposition being, of course, spherical dipole radiation). Near an interface, however, this mode structure is different, and therefore the coupling of the atomic dipole  $\underline{\mu}$  to this vacuum field will be different. Since this interaction is responsible for the spontaneous decay and the emission of fluorescence, the atom will behave differently near a medium. For instance, the atom cannot excite a plane-wave mode of the radiation field with a wave vector in the direction of the medium, without also partially exciting the mode with its wave vector in the specular direction.

We consider an atom, with dipole moment  $\underline{\mu}$ , which is positioned at  $\underline{r} = h\hat{e}_z$ ,  $h > 0$ , on the  $z$ -axis, and the plane  $z = 0$  separates the vacuum in  $z > 0$  from a nonlinear medium in  $z < 0$  (not necessarily half-infinite). Two strong counterpropagating laser beams with frequency  $\bar{\omega} > 0$  pump the medium (they travel parallel to the  $z = 0$  plane), such that a four-wave mixing process between the pumps and an external field, which is incident from the region  $z > 0$ , is responsible for the reflection of radiation back into the region  $z > 0$ . This 'device' is called a phase conjugator (PC). We shall not assume any

details about the four-wave mixing, nor about the linear properties of the medium, which keeps the theory very universal. It can be shown in general [18] that when a plane wave with wave vector  $\underline{k}$  and frequency  $\omega > 0$  is incident upon a PC, then there are two plane waves reflected back into the  $z > 0$  region. First there is of course the specular wave with wave vector  $\underline{k}_r$  and frequency  $\omega > 0$ , due to a dielectric constant which is unequal to unity. This radiation (when it comes from the dipole) is responsible for a change in lifetime of excited states, as described above. In addition, there is a phase-conjugated wave with wave vector  $\underline{k}_{pc} \approx \underline{k}$ , and this wave has the negative frequency

$$\omega' = -\omega - 2\bar{\omega} \quad (1.2)$$

Hence the phase-conjugated (pc) wave and the incident wave are almost counterpropagating, in contrast to the propagation direction of the specular wave (which we shall call the r-wave).

For pure plane waves it does not seem to make much of a difference in which direction the produced wave emanates from the medium: it is again a plane wave. Dipole radiation is a spherically-diverging wave, and for reflection at a linear medium (as r-waves) the total reflected field is again a diverging spherical wave. The phase-conjugated field (pc-waves), however, is a converging wave which is focused almost exactly on the radiating atom, because every Fourier plane-wave component retraces its path after reflection. Most r-wave photons are scattered away by the medium, but every pc-wave photon returns to the atom and can again interact with it. Such a process can therefore be anticipated to give rise to much stronger effects on atomic lifetimes and on the time evolution of the atomic density operator, as

compared to the influence of r-wave photons. As it turns out, however, the situation is more subtle, and simple interpretations as given above for linear reflection have to be modified carefully in order to picture a consistent interpretation of the behavior of an atom near a PC.

## II. RELAXATION

The interaction of the atomic dipole  $\underline{\mu}$  with the electromagnetic field of the vacuum gives rise to both spontaneous decay and the emission of fluorescence, and energy conservation obviously requires a relation between both processes. Elsewhere [19] we have derived an expression for the fluorescence radiation field, as it is emitted by the atom near the PC, and the result was expressed in terms of  $\underline{\mu}(t)$ , where the  $t$ -dependence signifies the Heisenberg picture. In evaluating quantum expectation values of certain field properties (like the intensity of emission, or the spectral distribution of the fluorescence), we can always transform to the Schrödinger picture in which the time evolution is governed entirely by the time dependence of the atomic density operator  $\rho_a(t)$ . Spontaneous decay amounts to a relaxation of  $\rho_a(t)$ , and in this way is the temporal evolution of the radiation field determined by the time dependence of the atomic density operator. In this paper we study the equation of motion for  $\rho_a(t)$ , and a combination with the results from our previous paper then completely determines, in principle, the temporal and spectral properties of phase-conjugated fluorescence.

Since the atom and the radiation field interact, we have to describe their joint quantum state by a single density operator  $\rho(t)$ . However, we are only interested in the state of the atom, irrespective of the state of the field. This atomic state is defined by



$$\rho_a(t) = \text{Tr}_r \rho(t) \quad , \quad (2.1)$$

where the trace runs over the states of the field only. In general, the time evolution of  $\rho(t)$  is given by

$$i\hbar \frac{d\rho}{dt} = [H, \rho] \quad , \quad (2.2)$$

and the Hamiltonian has the form

$$H = H_a + H_r + H_{ar} \quad , \quad (2.3)$$

in obvious notation. Then it is a standard procedure [20-22] to derive an equation of motion for  $\rho_a(t)$  from Eq. (2.2), under the condition that the radiation field can be considered as a thermal reservoir, which is no approximation at all for the present problem. Also, we make the Markov approximation, which is perfectly justified for the interaction between a dipole and the vacuum radiation field. We shall adopt the compact Liouville notation, since this appears to be the most convenient framework to describe relaxation phenomena [23]. With the definition of the Liouvillians  $L_i$

$$L_i \sigma = \hbar^{-1} [H_i, \sigma] \quad , \quad i = a, r, ar \quad , \quad (2.4)$$

where  $\sigma$  is an arbitrary operator in Hilbert space, the equation of motion reads

$$i \frac{d\rho}{dt} = (L_a + L_r + L_{ar}) \rho \quad . \quad (2.5)$$

From Eq. (2.5) one then derives the equation of motion for  $\rho_a(t)$ ,

$$i \frac{d\rho_a}{dt} = (L_a - i\Gamma)\rho_a, \quad (2.6)$$

where the relaxation operator  $\Gamma$  is found to be

$$\Gamma\sigma_a = \text{Tr}_r L_{ar} \int_0^\infty d\tau e^{-i(L_a + L_r)\tau} L_{ar} e^{iL_a\tau} (\sigma_a \bar{\rho}_r). \quad (2.7)$$

This relation defines the action of  $\Gamma$  on an arbitrary atomic operator  $\sigma_a$ , and it involves the thermal-equilibrium density operator  $\bar{\rho}_r$  of the radiation field. For the present situation we have

$$\bar{\rho}_r = |0\rangle\langle 0|, \quad (2.8)$$

which indicates that the electromagnetic field is in the vacuum state. In Eq. (2.6), the Liouvillian  $L_a$  represents the free evolution of the atom, as it would be without the coupling to the radiation field, and the relaxation operator  $\Gamma$  accounts for spontaneous decay. Its matrix elements are related to the lifetimes of atomic states, and they include the time evolution of atomic coherences. The purpose of this paper is to evaluate  $\Gamma$ , as it is given by Eq. (2.7), for an atom near a PC, and to interpret the physical behavior of the atom from the solution of Eq. (2.6).

### III. DIPOLE COUPLING

In order to evaluate expression (2.7) for  $\Gamma$ , we need to specify the interaction Hamiltonian  $H_{ar}$ . We write  $E_v(\underline{r}, t)$  for the electric-field operator

of the vacuum, and we take the Schrödinger and Heisenberg representation to coincide at  $t = 0$ . With  $\underline{h}$  the position of the atom, the interaction Hamiltonian in the dipole approximation then reads

$$H_{ar} = -\underline{\mu} \cdot \underline{E}_v(\underline{h}, 0) \quad . \quad (3.1)$$

The explicit form of  $\underline{E}_v(\underline{r}, t)$  is irrelevant for the remainder of this section. When we substitute expression (3.1) into Eq. (2.7) and work out the commutators which define the various Liouvillians, then it appears that  $\Gamma$  can be represented by the simpler form

$$\Gamma\sigma = \sum_i [\mu_i, Q_i\sigma - \sigma Q_i^\dagger] \quad , \quad (3.2)$$

where the summation runs over the Cartesian coordinates  $i = x, y, z$ . Here we have dropped the subscript 'a' on the atomic operator  $\sigma$ , as we shall do in the remainder of the paper. Also, we shall write  $\rho$  for the density operator, instead of  $\rho_a$ . The Hilbert-space operator  $Q_i$  (not a Liouvillian) is defined as

$$Q_i = \int_0^\infty d\tau e^{-iL_a\tau} \sum_j f_{ij}(\tau) \mu_j \quad , \quad (3.3)$$

in terms of the nine functions  $f_{ij}(\tau)$ . These quantities are the vacuum correlation functions, which are given by

$$f_{ij}(\tau) = \frac{1}{\hbar^2} \langle 0 | E_v(\underline{h}, \tau)_i E_v(\underline{h}, 0)_j | 0 \rangle \quad , \quad (3.4)$$

and where we have used Eq. (2.8) for  $\bar{\rho}_r$ . Notice that the right-hand side of Eq. (3.2) does not contain field Liouvillians anymore, and that the operators  $Q_i$  only involve the Liouvillian  $L_a$  of the free evolution and the Cartesian components  $\mu_j$  of the dipole operator. All necessary information about the vacuum radiation field is contained in the nine correlation functions  $f_{ij}(r)$ .

#### IV. PLANE-WAVE MODES

Before we write down the expression for the vacuum field  $F(\underline{r}, t)$ , we briefly summarize the fundamental plane-wave mode solution for a PC, mainly to set up the notation. The nonlinear medium is assumed to occupy the region  $0 > z > -\Delta$ , with  $\Delta > 0$  the layer thickness. Then we consider a monochromatic polarized plane-wave as the incident field from the region  $z > 0$ , with positive-frequency part

$$\underline{E}_{\text{inc}}(\underline{r}, t)^{(+)} = E_{\underline{k}\sigma} e_{\underline{k}\sigma} e^{i(\underline{k} \cdot \underline{r} - \omega t)} \quad (4.1)$$

Here,  $\omega = ck > 0$  and  $\underline{k}$  is assumed to be real (travelling wave). For later purposes we allow the amplitude factor  $E_{\underline{k}\sigma}$  to be a quantum operator. For the unit polarization vector  $e_{\underline{k}\sigma}$  we take either  $\sigma = s$  or  $\sigma = p$ , corresponding to a surface-polarized and a plane-polarized wave, respectively. Then it can be shown [18,24] that the reflected waves, the transmitted waves, and the waves inside the medium are also plane waves, and they have the same polarization  $\sigma$  as the incident wave. Also, when we write for the wave vector of the incident field

$$\underline{k} = \underline{k}_{\parallel} + k_{z-z} \underline{e}_z \quad (4.2)$$

where the subscript  $\parallel$  refers to the plane  $z = 0$ , then every other wave must have a wave vector with the same parallel component  $k_{\parallel}$ , due to the boundary conditions at  $z = 0$  and  $z = -\Delta$ . Therefore, every wave vector can be written as

$$\underline{k}_i = k_{\parallel} + k_{i,z} \underline{e}_z, \quad (4.3)$$

and the value of  $k_{i,z}$  is determined (up to its sign) by the dispersion relation for either the medium or the vacuum. For a wave in vacuum with wave vector  $\underline{k}_i$  we take the unit polarization vectors as

$$\underline{e}_{k_i s} = \frac{1}{k_{\parallel}} k_{\parallel} \times \underline{e}_z, \quad (4.4)$$

$$\underline{e}_{k_i p} = \frac{1}{k_i} k_i \times \underline{e}_{k_i s}. \quad (4.5)$$

The waves in the medium are in general not transverse, and the expressions for their polarization vectors are more complicated [24]. For the present problem, however, we need only the waves in vacuum, so that we shall not write down the form of the fields in the medium.

For given values of  $k = \omega/c$  and  $k_{\parallel}$  (or the angle of incidence), the  $z$ -component of the wave vector of the incident field is

$$k_z = -\sqrt{k^2 - k_{\parallel}^2}. \quad (4.6)$$

As mentioned in the Introduction, there are in general two waves reflected back into the region  $z > 0$ , and they have wave vectors  $\underline{k}_r$  and  $\underline{k}_{pc}$ . From the

dispersion relation, and the fact that both waves must travel into the positive  $z$ -direction, we find the  $z$ -components of these wave vectors to be

$$k_{r,z} = -k_z, \quad (4.7)$$

$$k_{pc,z} = -\sqrt{\rho^2 k^2 - k_{\parallel}^2}, \quad (4.8)$$

where we introduced the dimensionless detuning parameter

$$\rho = -\omega'/\omega, \quad (4.9)$$

in terms of  $\omega'$  from Eq. (1.2). Then the positive-frequency part of the total field in  $z > 0$  assumes the form

$$\begin{aligned} \underline{E}(\underline{r}, t)^{(+)} = & \underline{E}_{k\sigma} e^{i(\underline{k} \cdot \underline{r} - \omega t)} + \underline{R}_{k\sigma - k_r \sigma} e^{i(\underline{k}_r \cdot \underline{r} - \omega t)} \\ & + \underline{E}_{k\sigma}^{\dagger} \underline{P}_{k\sigma - k_{pc} \sigma}^* e^{-i(\underline{k}_{pc} \cdot \underline{r} + \rho \omega t)}, \end{aligned} \quad (4.10)$$

in terms of the Fresnel reflection coefficients  $\underline{R}_{k\sigma}$  and  $\underline{P}_{k\sigma}$  for the  $r$ -wave and the  $pc$ -wave, respectively. These Fresnel coefficients contain detailed information about the linear interaction, the four-wave mixing process, the layer thickness, etc. They can be found by solving Maxwell's equations for the given incident wave, although the result depends in a very complicated way on the various parameters [18,24]. It is important to notice the dagger in the last term on the right-hand side of Eq. (4.10). For an incident field with frequency  $\omega > 0$ , the  $pc$ -wave is emitted at the negative frequency  $\omega'$ , and

consequently this is an  $\underline{E}^{(-)}$  field. From the general relation for an electric field

$$\underline{E}(\underline{r}, t)^{(+)} = (\underline{E}(\underline{r}, t)^{(-)})^\dagger, \quad (4.11)$$

which follows from the Hermiticity of  $\underline{E}$ , we then obtain a dagger on  $\underline{E}_{\underline{k}\sigma}$  (and a star on  $\underline{P}_{\underline{k}\sigma}$ ) in Eq. (4.10). For a classical field this is a complex conjugation, which is where the term 'phase conjugation' originates from. Notice that also the spatial part  $\exp(-i\underline{k}_{pc} \cdot \underline{r}) \approx \exp(-i\underline{k} \cdot \underline{r})$  is the complex conjugate of the spatial part of the incident field, apart from a small shift in wave number (due to  $\rho \neq 1$ ). In quantum mechanics this complex conjugation translates into a Hermitian conjugation, as is well known [25-27].

The incident wave travels through the layer and exits the medium at the interface  $z = -\Delta$ . This transmitted wave (t-wave) has wave vector  $\underline{k}_t$ , and obviously we must have

$$\underline{k}_t = \underline{k}, \quad (4.12)$$

whereas the frequency of the wave equals  $\omega$ . When this incident wave hits the boundary  $z = -\Delta$ , then a part of it will be reflected back into the medium, and approximately into the specular direction, just as in linear optics. Inside the medium this positive-frequency wave will couple to a counterpropagating negative-frequency wave, due to the nonlinear interaction, and this wave exits the medium at the boundary  $z = -\Delta$ . We call this the nonlinear wave (nl-wave), and its wave vector  $\underline{k}_{nl}$  is approximately equal to  $\underline{k}_r$ . Taking into account the frequency mismatch, we find for the z-component of  $\underline{k}_{nl}$

$$k_{nl,z} = \sqrt{\rho^2 k^2 - k_{\parallel}^2} = -k_{pc,z} \quad (4.13)$$

The total positive-frequency field in  $z < -\Delta$  then becomes

$$\begin{aligned} \underline{E}(\underline{r}, t)^{(+)} = & \underline{E}_{k\sigma} T_{k\sigma} e_{k\sigma} e^{i(\underline{k} \cdot \underline{r} - \omega t)} \\ & + \underline{E}_{k\sigma}^{\dagger} N_{k\sigma}^* e_{k_{nl}\sigma} e^{-i(\underline{k}_{nl} \cdot \underline{r} + \rho \omega t)} \end{aligned} \quad (4.14)$$

in terms of the Fresnel transmission coefficients  $T_{k\sigma}$  and  $N_{k\sigma}$  for the t-wave and nl-wave, respectively. Notice that we label all Fresnel coefficients with the wave vector  $\underline{k}$  of the incident wave, rather than with the wave vector of the corresponding wave. This will turn out to be most practical later on.

For a given incident plane wave we find a total of five plane waves outside the medium: three in  $z > 0$  and two in  $z < -\Delta$ . It is easy to show that this is the most general solution of Maxwell's equations (or Heisenberg's equations for quantum fields), due to the restrictions put on the wave numbers by the dispersion relation in vacuum, by the causality requirement that all waves must travel away from the medium, and by the phase-matching conditions in  $z = 0$  and  $z = -\Delta$ . In the same way it can be shown that the most general solution for the waves inside the medium consists of four sets of two counterpropagating waves. This brings the total count of the number of waves to thirteen, which are all present due to the excitation by a single incident plane wave. By matching these waves across the boundaries at  $z = 0$  and  $z = -\Delta$ , we can derive explicit expressions for the Fresnel coefficients [24]. In this paper we shall consider these coefficients as given parameters of the problem. Figure 1 illustrates the situation.



## V. VACUUM FIELD

With the plane-wave solution from the previous section, we can now construct the explicit form of the vacuum field  $\underline{E}_v(\underline{r}, t)$ , and because the dipole which couples to this field is situated in  $z > 0$ , we shall consider  $\underline{E}_v$  in this region only. To this end, we first recall that the positive-frequency part of the electric-field operator in empty space is given by

$$\underline{E}_e(\underline{r}, t)^{(+)} = \sum_{\underline{k}\sigma} \sqrt{\frac{\hbar\omega}{2\epsilon_0 V}} a_{\underline{k}\sigma} \underline{e}_{\underline{k}\sigma} e^{i(\underline{k}\cdot\underline{r} - \omega t)} \quad (5.1)$$

with  $\omega = ck$ ,  $V$  the quantization volume, and  $a_{\underline{k}\sigma}$  the annihilation operator for a photon in the mode  $\underline{k}\sigma$ . For the unit polarization vectors  $\underline{e}_{\underline{k}\sigma}$  we take  $\underline{e}_{\underline{k}s}$  and  $\underline{e}_{\underline{k}p}$  from Eqs. (4.4) and (4.5), respectively. Then we introduce the operators

$$\underline{E}_{\underline{k}\sigma} = \sqrt{\frac{\hbar\omega}{2\epsilon_0 V}} a_{\underline{k}\sigma} \quad (5.2)$$

which gives

$$\underline{E}_e(\underline{r}, t)^{(+)} = \sum_{\underline{k}\sigma} \underline{E}_{\underline{k}\sigma} \underline{e}_{\underline{k}\sigma} e^{i(\underline{k}\cdot\underline{r} - \omega t)} \quad (5.3)$$

This shows that  $\underline{E}_e(\underline{r}, t)^{(+)}$  is a summation over  $\underline{k}\sigma$  of plane waves which have exactly the form (4.1) of an incident monochromatic plane wave. Since the plane-wave solution from the previous section is the only one admitted by the Maxwell-Heisenberg equations, we conclude that every  $\underline{k}\sigma$ -term in Eq. (5.3) must give rise to an excitation of a fundamental plane-wave mode.

In Sec. IV we considered a plane wave which was incident from the region  $z > 0$ , corresponding to a negative value of  $k_z$ . In the summation (5.3), half the number of wave vectors  $\underline{k}$  has a negative  $z$ -component, and for these modes we can simply take Eq. (4.10), summed over  $\underline{k}\sigma$ , for the field in  $z > 0$ . The other half of the terms in Eq. (5.3) have  $k_z > 0$ , and they correspond to incident waves from the region  $z < -\Delta$ . In  $z > 0$ , these waves generate  $t$ -waves and  $nl$ -waves in analogy to Eq. (4.14). Therefore, the vacuum field in  $z > 0$  is

$$\begin{aligned}
 E_v(\underline{r}, t) = & \sum'_{\underline{k}\sigma} E_{\underline{k}\sigma} e_{\underline{k}\sigma} e^{i(\underline{k} \cdot \underline{r} - \omega t)} + R_{\underline{k}\sigma} e_{\underline{k}_r\sigma} e^{i(\underline{k}_r \cdot \underline{r} - \omega t)} \\
 & + \sum'_{\underline{k}\sigma} E_{\underline{k}\sigma}^\dagger P_{\underline{k}\sigma}^* e_{\underline{k}_{pc}\sigma} e^{-i(\underline{k}_{pc} \cdot \underline{r} + \rho\omega t)} \\
 & + \sum''_{\underline{k}\sigma} E_{\underline{k}\sigma} T'_{\underline{k}\sigma} e_{\underline{k}\sigma} e^{i(\underline{k} \cdot \underline{r} - \omega t)} \\
 & + \sum''_{\underline{k}\sigma} E_{\underline{k}\sigma}^\dagger N_{\underline{k}\sigma}'^* e_{\underline{k}_{nl}\sigma} e^{-i(\underline{k}_{nl} \cdot \underline{r} + \rho\omega t)} + \text{H.c.} , \quad (5.4)
 \end{aligned}$$

where a prime (double prime) on a summation sign indicates that the summation runs over  $\underline{k}$  values with  $k_z < 0$  ( $k_z > 0$ ) only. The summations on the right-hand side of Eq. (5.4) form together the positive-frequency part of the field, and the addition of the Hermitian conjugate then yields the total vacuum field. The primes on  $T'_{\underline{k}\sigma}$  and  $N_{\underline{k}\sigma}'^*$  are a reminder that these are the Fresnel

coefficients for a wave which is incident from the region  $z < -\Delta$ , and is identical in form to expression (4.1). The absolute value of  $T$  and  $T'$  is of course the same when both incident waves have the same frequency, angle of incidence, and polarization, but their phase will be different. For a transparent medium (unit dielectric constant) the  $r$ -waves and  $nl$ -waves disappear to a good approximation, which gives  $R_{\underline{k}\sigma} = N'_{\underline{k}\sigma} = 0$  in Eq. (5.4). The  $t$ -waves in  $z > 0$  remain present in this limit, and they were called 'quantum noise' by Gaeta and Boyd [28], following Caves [29]. For a transparent medium, and without the nonlinear interaction, we obviously have  $T'_{\underline{k}\sigma} = 1$  (because then  $\underline{E}_v = \underline{E}_e$ ). It can be shown [24,30] that the nonlinear interaction always gives  $|T'_{\underline{k}\sigma}| > 1$ , provided that the dielectric constant is unity.

## VI. VACUUM CORRELATION FUNCTIONS

With the explicit expression (5.4) for the vacuum field, we can now evaluate the vacuum correlation functions  $f_{ij}(\underline{r})$  from Eq. (3.6). We set  $\underline{r} = \underline{h}$ , and  $\tau = \tau$  and  $\tau = 0$  for the  $i$  and the  $j$  component of  $\underline{E}_v$ , respectively. Multiplying both expressions and taking the vacuum expectation value then yields

$$\begin{aligned}
f_{ij}(\tau) = & \sum_{\underline{k}\sigma} \frac{\omega}{2\epsilon_0 \hbar V} \{ P_{\underline{k}\sigma} e_{\underline{k}_{pc}\sigma} e^{i(\underline{k}_{pc} \cdot \underline{h} + \rho\omega\tau)} \\
& + (e_{\underline{k}\sigma} e^{i\underline{k} \cdot \underline{h}} + R_{\underline{k}\sigma} e_{\underline{k}_r\sigma} e^{i\underline{k}_r \cdot \underline{h}}) e^{-i\omega\tau} \}_i \\
& \times \{ P_{\underline{k}\sigma}^* e_{\underline{k}_{pc}\sigma} e^{-i\underline{k}_{pc} \cdot \underline{h}} + (e_{\underline{k}\sigma} e^{-i\underline{k} \cdot \underline{h}} + R_{\underline{k}\sigma}^* e_{\underline{k}_r\sigma} e^{-i\underline{k}_r \cdot \underline{h}}) \}_j \\
& + \sum_{\underline{k}\sigma} \frac{\omega}{2\epsilon_0 \hbar V} \{ N'_{\underline{k}\sigma} e_{\underline{k}_{nl}\sigma} e^{i(\underline{k}_{nl} \cdot \underline{h} + \rho\omega\tau)} + e_{\underline{k}\sigma} T'_{\underline{k}\sigma} e^{i(\underline{k} \cdot \underline{h} - \omega\tau)} \}_i \\
& \times \{ N_{\underline{k}\sigma}'^* e_{\underline{k}_{nl}\sigma} e^{-i\underline{k}_{nl} \cdot \underline{h}} + e_{\underline{k}\sigma} T_{\underline{k}\sigma}'^* e^{-i\underline{k} \cdot \underline{h}} \}_j . \quad (6.1)
\end{aligned}$$

Next we have to substitute the values of the various wave vectors, as they were introduced in Sec. IV. We notice that all wave vectors in the exponents appear in the combination  $\underline{k}_i \cdot \underline{h} = \hbar k_{i,z}$ , so that we only need their  $z$ -components. At this stage we make a slight approximation. The Fresnel coefficients  $P_{\underline{k}\sigma}$  and  $N'_{\underline{k}\sigma}$  only deviate considerably from zero when the frequency  $\omega$  of the incident wave is in close resonance with the pump frequency  $\bar{\omega}$ . Therefore, in factors multiplying these Fresnel coefficients we can safely replace  $\omega$  by  $\bar{\omega}$ . This amounts to the approximations  $k_{pc,z} \approx k_z$ ,  $k_{nl,z} \approx -k_z$ ,  $e_{\underline{k}_{pc}\sigma} \approx e_{\underline{k}\sigma}$  and  $e_{\underline{k}_{nl}\sigma} \approx e_{\underline{k}_r\sigma}$ , but the full resonant behavior of the Fresnel coefficients as a function of  $\omega$  will be retained. Then we obtain

$$\begin{aligned}
f_{ij}(\tau) = & \sum_{\underline{k}\sigma} \frac{\omega}{2\epsilon_0 \hbar V} \{ P_{\underline{k}\sigma} e_{\underline{k}\sigma} e^{i\rho\omega\tau} + (e_{\underline{k}\sigma} + R_{\underline{k}\sigma} e_{\underline{k}_r\sigma} e^{-2i\underline{k}\cdot\underline{h}}) e^{-i\omega\tau} \}_i \\
& \times ((P_{\underline{k}\sigma}^* + 1) e_{\underline{k}\sigma} + R_{\underline{k}\sigma}^* e_{\underline{k}_r\sigma} e^{2i\underline{k}\cdot\underline{h}})_j \\
& + \sum_{\underline{k}\sigma} \frac{\omega}{2\epsilon_0 \hbar V} \{ N'_{\underline{k}\sigma} e_{\underline{k}_r\sigma} e^{-2i\underline{k}\cdot\underline{h}} e^{i\rho\omega\tau} + T'_{\underline{k}\sigma} e_{\underline{k}\sigma} e^{-i\omega\tau} \}_i \\
& \times (N'^*_{\underline{k}\sigma} e_{\underline{k}_r\sigma} e^{2i\underline{k}\cdot\underline{h}} + T'^*_{\underline{k}\sigma} e_{\underline{k}\sigma})_j .
\end{aligned} \tag{6.2}$$

Notice that we have not set  $\rho = 1$  in  $\exp(i\rho\omega\tau)$ , since this would affect the  $\tau$ -dependence of  $f_{ij}(\tau)$ .

As the next step we want to perform the summations over  $\underline{k}$ , with the help of the identity

$$\frac{1}{V} \sum'_{\underline{k}} (\dots) = \frac{1}{8\pi^3} \int_{k_z < 0} d^3\underline{k} (\dots) , \tag{6.3}$$

and similarly for the double-prime summation. The integral is most easily carried out in spherical coordinates, for which we have

$$\frac{1}{V} \sum'_{\underline{k}} (\dots) = \frac{1}{8\pi^3} \int_0^\infty dk \int_{\pi/2}^\pi d\theta \int_0^{2\pi} d\phi k^2 \sin\theta (\dots) , \tag{6.4}$$

and for the summation over  $k_z > 0$  we replace the integration limits  $(\pi/2, \pi)$  for the  $\theta$ -integral by  $(0, \pi/2)$ . For the  $\underline{k}$ -vector we have

$$\underline{k} = k \underline{e}_r, \quad (6.5)$$

and with Eqs. (4.4) and (4.5) we find for the polarization vectors

$$\underline{e}_{ks} = \underline{e}_{kr}s = -\underline{e}_\phi, \quad (6.6)$$

$$\underline{e}_{kp} = \underline{e}_\theta, \quad (6.7)$$

$$\underline{e}_{krp} = -\underline{e}_\theta - 2\sin\theta \underline{e}_z, \quad (6.8)$$

in terms of the standard spherical unit vectors  $\underline{e}_r$ ,  $\underline{e}_\theta$  and  $\underline{e}_\phi$ .

When we make the substitutions (6.4)-(6.8) into Eq. (6.2) then we get a very lengthy expression for  $f_{ij}(\tau)$ , and for every one of the nine combinations  $(i,j)$  this expression is different. A great simplification arises from the fact that the Fresnel coefficients only depend on  $k$  and  $\theta$  (or the frequency of the incident wave and the angle of incidence), but not on  $\phi$ . Therefore, the only  $\phi$ -dependence of the integrand enters through the polarization vectors from Eqs. (6.6)-(6.8), and the integration over  $\phi$  can be performed immediately in each of the nine combinations. We then find that this integral equals zero whenever  $i \neq j$ , and that  $i = j = x$  gives the same result as  $i = j = y$ . Consequently, we can write

$$f_{ij}(\tau) = \delta_{ij} f_{ii}(\tau), \quad (6.9)$$

and for the three nonzero correlation functions we have

$$f_{\parallel}(\mathbf{r}) = f_{xx}(\mathbf{r}) = f_{yy}(\mathbf{r}) \quad , \quad (6.10)$$

$$f_{\perp}(\mathbf{r}) = f_{zz}(\mathbf{r}) \quad , \quad (6.11)$$

which defines  $f_{\parallel}(\mathbf{r})$  and  $f_{\perp}(\mathbf{r})$ . It can be shown [31] that results (6.9)-(6.11) are not just a lucky simplification for the present problem, but that these relations can be derived in general from the invariance of the vacuum for rotations about the z-axis, and for reflections in a plane through the z-axis. For a spherically-symmetric vacuum (like empty space), we find in addition that  $f_{\perp}(\mathbf{r})$  must necessarily equal  $f_{\parallel}(\mathbf{r})$ .

For the integration over  $\theta$  we change the integration variable to the angle of incidence  $\theta_i$  for a given value of  $\underline{k}$ , and the relation between  $\theta_i$  and  $\theta$  is

$$\theta_i = \begin{cases} \pi - \theta & , \quad \text{for } k_z < 0 \\ \theta & , \quad \text{for } k_z > 0 \end{cases} \quad . \quad (6.12)$$

Then we know that Fresnel coefficients only depend on  $\theta_i$  through  $\cos\theta_i$  (or  $\sin\theta_i$ ), because their angle-of-incidence dependence is purely geometrical and can be expressed in cross and dot products between the various wave vectors, polarization vectors and  $\underline{e}_z$ . We write

$$P_{\underline{k}\sigma} = P_{\sigma}(\omega, \cos\theta_i) \quad , \quad (6.13)$$

with  $\omega > 0$  the frequency of the incident wave, and similarly for the other Fresnel coefficients. For the  $k$ - and  $\theta_i$ -integration we then change integration variables according to  $\omega = ck$  and  $u = \cos\theta_i$ , respectively. For the two vacuum correlation functions we then obtain

$$f_{\perp}(\tau) = \frac{1}{8\pi^2 \epsilon_0 \hbar c^3} \int_0^\infty d\omega \omega^3 \int_0^1 du (1 - u^2) \\ \times \left\{ (P_p e^{i\rho\omega\tau} + (1 + R_p e^{2i\hbar\omega u/c}) e^{-i\omega\tau}) (P_p^* + 1 + R_p^* e^{-2i\hbar\omega u/c}) \right. \\ \left. + (N_p' e^{-2i\hbar\omega u/c} e^{i\rho\omega\tau} + T_p' e^{-i\omega\tau}) (N_p'^* e^{2i\hbar\omega u/c} + T_p'^*) \right\}, \quad (6.14)$$

$$f_{\parallel}(\tau) = \frac{1}{16\pi^2 \epsilon_0 \hbar c^3} \int_0^\infty d\omega \omega^3 \int_0^1 du \\ \times \left\{ (P_s e^{i\rho\omega\tau} + (1 + R_s e^{2i\hbar\omega u/c}) e^{-i\omega\tau}) (P_s^* + 1 + R_s^* e^{-2i\hbar\omega u/c}) \right. \\ \left. + (N_s' e^{-2i\hbar\omega u/c} e^{i\rho\omega\tau} + T_s' e^{-i\omega\tau}) (N_s'^* e^{2i\hbar\omega u/c} + T_s'^*) \right\} \\ + \frac{1}{16\pi^2 \epsilon_0 \hbar c^3} \int_0^\infty d\omega \omega^3 \int_0^1 du u^2 \\ \times \left\{ (P_p e^{i\rho\omega\tau} + (1 - R_p e^{2i\hbar\omega u/c}) e^{-i\omega\tau}) (P_p^* + 1 - R_p^* e^{-2i\hbar\omega u/c}) \right. \\ \left. + (N_p' e^{-2i\hbar\omega u/c} e^{i\rho\omega\tau} - T_p' e^{-i\omega\tau}) (N_p'^* e^{2i\hbar\omega u/c} - T_p'^*) \right\}, \quad (6.15)$$



where all Fresnel coefficients have  $(\omega, u)$  as their arguments. These coefficients depend in complicated way on  $\omega$  and  $u$ , in general, and it does not appear possible to evaluate the integrals in Eqs. (6.14) and (6.15).

## VII. LAPLACE TRANSFORM

The vacuum correlation functions  $f_{ij}(\tau)$  only enter the spontaneous-decay operator through  $Q_i$  from Eq. (3.5). When we expand  $\exp(iL_a \tau)$  onto the energy eigenstates of the atom, then this factor effectively turns into  $\exp(i\omega\tau)$  with  $\omega$  the energy separation between two levels, divided by  $\hbar$ . Consequently, the vacuum correlation functions only appear as their Laplace transform

$$\bar{f}_\alpha(\omega) = \int_0^\infty d\tau e^{i\omega\tau} f_\alpha(\tau) \quad , \quad \alpha = \perp, \parallel \quad , \quad (7.1)$$

where  $\omega$  can be either positive or negative. Since the factors in Eqs. (6.14) and (6.15) which depend on  $\tau$  are all exponentials, we can evaluate the Laplace transforms with the identity

$$\int_0^\infty d\tau e^{i(\omega \pm \omega')\tau} = \pi \delta(\omega \pm \omega') + P \frac{i}{\omega \pm \omega'} \quad , \quad (7.2)$$

where  $P$  stands for principle value. We shall omit the principle-value part, since it leads to a small level shift and not to spontaneous decay. After integration over  $\tau$  we can also perform the  $\omega$ -integration, which finally gives

$$\bar{f}_\alpha(\omega) = \begin{cases} \eta(\omega) \{x_\alpha(\omega) + z_\alpha(\omega)\} \quad , & \omega > 0 \\ \eta(2\bar{\omega} + \omega) \{y_\alpha(2\bar{\omega} + \omega) + z_\alpha(2\bar{\omega} + \omega)^*\} \quad , & \omega < 0 \end{cases} \quad , \quad (7.3)$$

with  $\alpha = \perp, \parallel$ . Here we introduced

$$\eta(\omega) = \frac{\omega^3}{6\pi\epsilon_0 \hbar c^3}, \quad (7.4)$$

and the six parameter functions

$$x_{\perp}(\omega) = \frac{3}{4} \int_0^1 du (1-u^2) (|1 + R_p e^{i\beta u}|^2 + |T'_p|^2), \quad (7.5)$$

$$x_{\parallel}(\omega) = \frac{3}{8} \int_0^1 du (|1 + R_s e^{i\beta u}|^2 + u^2 |1 - R_p e^{i\beta u}|^2 + |T'_s|^2 + u^2 |T'_p|^2), \quad (7.6)$$

$$y_{\perp}(\omega) = \frac{3}{4} \int_0^1 du (1-u^2) (|P_p|^2 + |N'_p|^2), \quad (7.7)$$

$$y_{\parallel}(\omega) = \frac{3}{8} \int_0^1 du (|P_s|^2 + u^2 |P_p|^2 + |N'_s|^2 + u^2 |N'_p|^2), \quad (7.8)$$

$$z_{\perp}(\omega) = \frac{3}{4} \int_0^1 du (1-u^2) (P_p^* + e^{i\beta u} (P_p^* R_p + T_p'^* N_p'^*)), \quad (7.9)$$

$$z_{\parallel}(\omega) = \frac{3}{8} \int_0^1 du (P_s^* + u^2 P_p^* + e^{i\beta u} (P_s^* R_s - u^2 P_p^* R_p + T_s'^* N_s'^* - u^2 T_p'^* N_p'^*)), \quad (7.10)$$

which are defined for  $\omega > 0$  only. All Fresnel coefficients have  $(\omega, u)$  as their argument, and the parameter  $\beta$  is defined by

$$\beta = \frac{2\omega h}{c} \quad (7.11)$$

Notice that the atom-surface distance  $h$  only enters via the parameter  $\beta$ , and therefore the two parameter functions  $y_{\perp}(\omega)$  and  $y_{\parallel}(\omega)$  are independent of this distance. Furthermore, the functions  $x_{\alpha}(\omega)$  and  $y_{\alpha}(\omega)$  are real and non-negative, but  $z_{\perp}(\omega)$  and  $z_{\parallel}(\omega)$  are complex-valued, in general.

### VIII. PARAMETER FUNCTIONS

Spontaneous decay of any atom near any linear or nonlinear medium occupying the region  $0 > z > -\Delta$  is determined completely by the two functions  $\tilde{f}_{\perp}(\omega)$  and  $\tilde{f}_{\parallel}(\omega)$ , which can be parametrized by the six functions  $x_{\alpha}(\omega)$ ,  $y_{\alpha}(\omega)$ ,  $z_{\alpha}(\omega)$ . These six parameter functions are again determined by the eight Fresnel coefficients  $R_{\sigma}$ ,  $P_{\sigma}$ ,  $T'_{\sigma}$ ,  $N'_{\sigma}$  ( $\sigma = s, p$ ), and the atom-surface distance parameter  $\beta$ . As soon as the Fresnel coefficients for a certain configuration are given as a function of the cosine of the angle of incidence  $u$ , then the integrals in Eqs. (7.5)-(7.10) can be evaluated, in principle. The dependence on the frequency  $\omega$  is only parametric, where  $\omega$  equals a Bohr frequency of the atom. In order to illuminate the significance of the parameter functions, we work out some important limiting cases.

#### A. *Empty space*

Without a medium we simply have an atom in  $\underline{r} = \underline{h}$ , and surrounded by empty space. Then the Fresnel coefficients obviously are

$$R_{\sigma} = P_{\sigma} = N'_{\sigma} = 0, \quad T'_{\sigma} = 1, \quad (8.1)$$

and the parameter functions are readily found to be

$$x_{\alpha} = 1, \quad y_{\alpha} = z_{\alpha} = 0. \quad (8.2)$$

They are the same for  $\alpha = \perp$  and  $\alpha = \parallel$ , which indicates the spherical symmetry of the vacuum, and they are independent of  $\omega$  (and of  $h$ ).

#### B. Linear medium

When the medium is a dielectric, a metal, a thin film or a substrate, or any composition for which reflection and refraction is induced by the first-order susceptibility only, then every plane wave has the same frequency as an incident plane wave, and consequently we have

$$P_{\sigma} - N'_{\sigma} = 0, \quad R_{\sigma} \neq 0, \quad T'_{\sigma} \neq 0, \quad (8.3)$$

in general. Then we find immediately

$$x_{\alpha} \neq 0, \quad y_{\alpha} = z_{\alpha} = 0, \quad (8.4)$$

and the vacuum correlation functions become

$$\tilde{f}_{\alpha}(\omega) = \begin{cases} \eta(\omega)x_{\alpha}(\omega), & \omega > 0 \\ 0, & \omega < 0 \end{cases}, \quad (8.5)$$

which will be different for the two values of  $\alpha$ . For positive frequencies,  $\tilde{f}_{\alpha}(\omega)$  is real and positive, but for negative frequencies  $\tilde{f}_{\alpha}(\omega)$  vanishes identically. Conversely,  $\tilde{f}_{\alpha}(\omega)$  can only acquire a finite value for  $\omega < 0$  due to a nonlinear process in the medium.

### C. Perfect conductor

The most simple and nontrivial linear medium is the perfect conductor (mirror), for which we have

$$T'_\sigma = 0 \quad , \quad R_s = -1 \quad , \quad R_p = 1 \quad , \quad (8.6)$$

in addition to Eq. (8.3). Then the integrals over  $u$  can be found analytically, and we get

$$x_\perp(\omega) = 1 - 3 \left\{ \frac{\cos\beta}{\beta^2} - \frac{\sin\beta}{\beta^3} \right\} \quad , \quad (8.7)$$

$$x_\parallel(\omega) = 1 - \frac{3}{2} \left\{ \frac{\sin\beta}{\beta} + \frac{\cos\beta}{\beta^2} - \frac{\sin\beta}{\beta^3} \right\} \quad , \quad (8.8)$$

which is a well-known result. It exhibits the famous oscillatory structure as a function of  $h$  (or  $\beta$ ), and in the limit  $h \rightarrow \infty$  we find  $x_\perp = x_\parallel = 1$ . This is the empty-space limit from subsection A, and it displays that far away from the surface the spontaneous decay is not affected anymore by the presence of the medium.

### D. Transparent PC

For most four-wave mixing PC's one uses a transparent crystal (unit dielectric constant), which has the advantage that the specular wave is (almost) absent and that the phase-conjugate reflectivity can be high. In principle, there is no limit on the value of  $P_\sigma$ , i.e.,

$$0 \leq |P_\sigma|^2 < \infty , \quad (8.9)$$

and for the specular waves we have

$$R_\sigma - N'_\sigma = 0 . \quad (8.10)$$

The t-waves are always there, and their Fresnel coefficients are related to  $P_\sigma$  according to

$$|T_\sigma|^2 - |P_\sigma|^2 = 1 , \quad (8.11)$$

where both  $T_\sigma$  and  $P_\sigma$  pertain to the same incident plane wave. It can be shown [24] that relation (8.11) holds for any angle of incidence and any frequency mismatch between  $\omega$  and  $\bar{\omega}$ . In particular, Eq. (8.11) implies

$$|T_\sigma|^2 \geq 1 , \quad (8.12)$$

indicating that the transmitted wave is always amplified in the four-wave mixer, as compared to the incident wave. Equation (8.9) allows for values of  $|P_\sigma|^2$  larger than unity, in which case also the pc-wave is stronger than the incident wave. Amplification in a four-wave mixing PC has been observed experimentally indeed [32,33]. It is also worth noting that relation (8.11) is responsible for the generation of two-photon coherent states in four-wave mixing [34,35].

For this most important and realistic example we find that  $x_\alpha$  and  $y_\alpha$  are related as

$$x_{\alpha}(\omega) = 1 + y_{\alpha}(\omega) \quad , \quad (8.13)$$

with

$$y_{\perp}(\omega) = \frac{3}{4} \int_0^1 du (1 - u^2) |P_p|^2 \quad , \quad (8.14)$$

$$y_{\parallel}(\omega) = \frac{3}{8} \int_0^1 du (|P_s|^2 + u^2 |P_p|^2) \quad , \quad (8.15)$$

where we have used Eq. (8.11) to eliminate  $|T_{\sigma}|^2$  in favor of  $|P_{\sigma}|^2$ . It should be noted that  $T'_{\sigma}$  from Eqs. (7.5) and (7.6) is not equal to  $T_{\sigma}$  in Eq. (8.11), although their absolute values are equal when they both pertain to the same angle of incidence. For  $z_{\alpha}$  we find

$$z_{\perp}(\omega) = \frac{3}{4} \int_0^1 du (1 - u^2) P_p^* \quad , \quad (8.16)$$

$$z_{\parallel}(\omega) = \frac{3}{8} \int_0^1 du (P_s^* + u^2 P_p^*) \quad . \quad (8.17)$$

In the expression for  $\tilde{f}_{\alpha}(\omega)$ , Eq. (7.3), we always get  $y_{\alpha}$  and  $z_{\alpha}$  in the combination  $y_{\alpha} + z_{\alpha}$ . From Eqs. (8.14) and (8.15) we see that  $y_{\alpha} \geq 0$ , and that also the integrand is non-negative for every value of  $u$ . Therefore, the contributions from all plane waves with different angles of incidence add up constructively. This in contrast to Eqs. (8.16) and (8.17) where the values of  $P_{\sigma}^*$  appear, rather than their absolute values. The Fresnel coefficients  $P_{\sigma}$  vary rapidly as a function of the angle of incidence [24], and in an oscillatory fashion around  $P_{\sigma} = 0$ . Integrated over the angle of incidence, as

in Eqs. (8.16) and (8.17), this will yield a very small value as compared to the integrated intensities  $|P_\sigma|^2$ . A safe approximation is therefore

$$z_\alpha(\omega) \approx 0, \quad (8.18)$$

when used in Eq. (7.3)

#### E. Ideal PC

When the values of  $|P_s|^2$  and  $|P_p|^2$  would be independent of the angle of incidence (and therefore equal to  $|P_s(\omega, u = 1)|^2$ , because  $|P_s| = |P_p|$  for  $u = 1$ ), then we could take them outside the integrals in Eqs. (8.14) and (8.15), which would give

$$y_\perp(\omega) = y_\parallel(\omega) = \frac{1}{2} |P|^2. \quad (8.19)$$

For illustrative purposes later on we define an ideal PC as a transparent PC for which, in addition, Eq. (8.19) holds. The reservoir correlation function then equals

$$\bar{F}_\alpha(\omega) = \begin{cases} \eta(\omega)(1 + \kappa|P|^2) & , \quad \omega > 0 \\ \kappa\eta(-\omega)|P|^2 & , \quad \omega < 0 \end{cases}, \quad (8.20)$$

both for  $\alpha = \perp$  and  $\alpha = \parallel$ . Furthermore we have assumed that  $|P_\sigma|$  is symmetric around  $\bar{\omega}$ , as a function of  $\omega$ , which is quite accurate in general. We like to



emphasize that Eq. (8.19) is not necessarily a good approximation, and also that  $|P|^2$  on the right-hand side should not be interpreted as the reflectivity at normal incidence. The value  $|P|^2$  can be viewed as an average of  $|P_\sigma|^2$  over the angle of incidence, where the 'average' is defined by Eqs. (8.14) and (8.15). The approximation is then that Eqs. (8.14) and (8.15) give comparable results. In an approximation where  $P_\sigma(\omega, u)$  is assumed to be independent of  $u$ , we would find in addition that  $z_\alpha(\omega) = \hbar P^*$ , according to Eqs. (8.16) and (8.17). It is easy to show that this leads to inconsistencies, and that the appropriate approximation is given by Eq. (8.18). Also note that  $\bar{f}_\alpha(\omega)$  is identical in form to the correlation function of a thermal photon reservoir with an average photon number of  $|P|^2/2$ .

#### IX. TWO LEVELS

With the explicit result for the vacuum correlation functions from Sec. VII, we can evaluate the spontaneous-decay operator  $\Gamma$  from Eq. (3.4) for any configuration of atomic levels. In this section we work out the general form of  $\Gamma$  for the case of two, possibly degenerate or nearly-degenerate, levels. With  $\hbar\omega_e$  and  $\hbar\omega_g$  as the energies of the excited state and ground state, respectively, the atomic Hamiltonian reads

$$H_a = \hbar\omega_e P_e + \hbar\omega_g P_g, \quad (9.1)$$

in terms of the projectors  $P_e$  and  $P_g$  on both states. The corresponding Liouvillian is

$$L_a \sigma = \omega_o [P_e, \sigma] \quad (9.2)$$

with  $\omega_0 = \omega_e - \omega_g$  as the transition frequency, and where we used the closure relation

$$P_e + P_g = 1 \quad (9.3)$$

For the calculation of  $Q_i$  from Eq. (3.5) we need the expansion of  $\exp(-iL_a r)$  in projectors

$$e^{-iL_a r} = \sum_{\alpha\beta} e^{i(\omega_\beta - \omega_\alpha)r} P_\alpha P_\beta, \quad (9.4)$$

which can be proven easily with  $P_\alpha^2 = P_\alpha$ ,  $\alpha = e, g$ . This exponential acts on the Cartesian component  $\mu_i$  of the dipole operator, which gives

$$e^{-iL_a r} \mu_i = e^{-i\omega_0 r} P_e \mu_i P_g + e^{i\omega_0 r} P_g \mu_i P_e, \quad (9.5)$$

and where we used

$$P_\alpha \mu P_\alpha = 0, \quad \alpha = e, g. \quad (9.6)$$

This relation follows from the fact that an atom cannot have a permanent dipole moment. Substitution of the right-hand side of Eq. (9.5) into Eq. (3.5) then yields

$$Q_i = \tilde{f}_{ii}(-\omega_0) P_e \mu_i P_g + \tilde{f}_{ii}(\omega_0) P_g \mu_i P_e. \quad (9.7)$$

For molecular transitions, Eq. (9.6) does not hold since a molecule can have a permanent dipole moment. Nevertheless, the expression for  $Q_i$  remains the same because the terms with  $P_{\alpha} \mu_i P_{\alpha}$  become proportional to  $\tilde{f}_{ii}(0)$ , which equals zero.

Then we substitute  $Q_i$  and its Hermitian conjugate into Eq. (3.4), work out the commutator, and drop the nonsecular terms,<sup>36</sup> which gives for  $\Gamma$

$$\begin{aligned} \Gamma \sigma = & \sum_i \{ \mu_i^2 [P_g \tilde{f}_{ii}(-\omega_o) + P_e \tilde{f}_{ii}(\omega_o)] \sigma \\ & + \sigma [P_g \tilde{f}_{ii}(-\omega_o)^* + P_e \tilde{f}_{ii}(\omega_o)^*] \mu_i^2 \\ & - 2\mu_i [P_g \sigma P_g \operatorname{Re} \tilde{f}_{ii}(-\omega_o) + P_e \sigma P_e \operatorname{Re} \tilde{f}_{ii}(\omega_o)] \mu_i \} \end{aligned} \quad (9.8)$$

Here we have used identities like

$$P_e \mu_i P_g = P_e \mu_i - \mu_i P_g, \quad (9.9)$$

which easily follow from Eqs. (9.3) and (9.6).

In Eq. (9.8) we have expressed  $\Gamma$  in terms of the Cartesian components  $\mu_i$  of the dipole operator and the projectors  $P_e$  and  $P_g$  on both levels, which is the most compact representation of  $\Gamma$ . For many purposes, however, it is more convenient to express  $\Gamma$  in atomic lowering and raising operators, rather than in projectors. The lowering and raising parts of  $\underline{\mu}$  are

$$\underline{\mu}^{(+)} = P_g \underline{\mu} P_e, \quad (9.10)$$

$$\underline{\mu}^{(-)} = P_e \underline{\mu} P_g, \quad (9.11)$$

respectively, and the total dipole moment is

$$\underline{\mu} = \underline{\mu}^{(+)} + \underline{\mu}^{(-)}. \quad (9.12)$$

Then we substitute its  $i$ -th component into Eq. (9.8) and use the notation with  $\perp$  and  $\parallel$ , which gives

$$\begin{aligned} \Gamma_0 = & \{ \tilde{f}_{\parallel}(-\omega_0) \underline{\mu}_{\parallel}^{(+)} \cdot \underline{\mu}_{\parallel}^{(-)} + \tilde{f}_{\parallel}(\omega_0) \underline{\mu}_{\parallel}^{(-)} \cdot \underline{\mu}_{\parallel}^{(+)} \\ & + \tilde{f}_{\perp}(-\omega_0) \underline{\mu}_{\perp}^{(+)} \cdot \underline{\mu}_{\perp}^{(-)} + \tilde{f}_{\perp}(\omega_0) \underline{\mu}_{\perp}^{(-)} \cdot \underline{\mu}_{\perp}^{(+)} \} \sigma \\ & + \sigma \{ \tilde{f}_{\parallel}(-\omega_0) \star \underline{\mu}_{\parallel}^{(+)} \cdot \underline{\mu}_{\parallel}^{(-)} + \tilde{f}_{\parallel}(\omega_0) \star \underline{\mu}_{\parallel}^{(-)} \cdot \underline{\mu}_{\parallel}^{(+)} \\ & + \tilde{f}_{\perp}(-\omega_0) \star \underline{\mu}_{\perp}^{(+)} \cdot \underline{\mu}_{\perp}^{(-)} + \tilde{f}_{\perp}(\omega_0) \star \underline{\mu}_{\perp}^{(-)} \cdot \underline{\mu}_{\perp}^{(+)} \} \\ & - 2 \sum_{i=x,y} \mu_i^{(-)} \sigma \mu_i^{(+)} \text{Re } \tilde{f}_{\parallel}(-\omega_0) - 2 \sum_{i=x,y} \mu_i^{(+)} \sigma \mu_i^{(-)} \text{Re } \tilde{f}_{\parallel}(\omega_0) \\ & - 2 \mu_z^{(-)} \sigma \mu_z^{(+)} \text{Re } \tilde{f}_{\perp}(-\omega_0) - 2 \mu_z^{(+)} \sigma \mu_z^{(-)} \text{Re } \tilde{f}_{\perp}(\omega_0) \quad (9.13) \end{aligned}$$

We notice that every term is proportional to  $\eta(\omega_0)$  from Eq. (7.4), but since there is only one transition frequency for a two-level system we simply write  $\eta$ .

# X. TWO-STATE ATOM AND IDEAL PC

In order to discover the physical mechanisms of spontaneous decay near a PC, and their relation to fluorescent emission, we consider the model case of a two-state atom, in combination with the ideal PC for which the vacuum correlation function is given by Eq. (8.20). With  $|e\rangle$  and  $|g\rangle$  as the wave functions of the excited state and ground state, respectively, the projectors on these states are

$$P_{\alpha} = |\alpha\rangle\langle\alpha| \quad , \quad \alpha = e, g \quad . \quad (10.1)$$

The raising part of the dipole operator can be written as

$$\underline{\mu}^{(-)} = \underline{\mu}_{eg} d \quad , \quad (10.2)$$

in terms of the matrix element  $\underline{\mu}_{eg} = \langle e | \underline{\mu} | g \rangle$  and the operator

$$d = |e\rangle\langle g| \quad , \quad (10.3)$$

whereas the lowering part is the Hermitian conjugate of  $\underline{\mu}^{(-)}$ . The Einstein coefficient for spontaneous decay in empty space is for this two-state atom given by Eq. (1.1), and can be written as

$$A = 2\eta |\langle e | \underline{\mu} | g \rangle|^2 \quad . \quad (10.4)$$

Then expression (9.13) for  $\Gamma$  can be simplified considerably, and in its most transparent form it reads

$$\begin{aligned}
\Gamma\sigma = & \frac{1}{2} A_e (P_e \sigma + \sigma P_e - 2d^\dagger \sigma d) \\
& + \frac{1}{2} A_g (P_g \sigma + \sigma P_g - 2d \sigma d^\dagger) .
\end{aligned} \tag{10.5}$$

Here we have introduced the real and positive parameters

$$A_e = A(1 + \frac{1}{2}|P|^2) , \tag{10.6}$$

$$A_g = \frac{1}{2} A|P|^2 , \tag{10.7}$$

whose significance will become clear in due course.

The equation of motion for the atomic density operator is given by Eq. (2.6) (with  $\rho_a \rightarrow \rho$ ). Most interesting is the time evolution of the populations of the two levels, defined by

$$n_\alpha(t) = \text{Tr } P_\alpha \rho(t) , \quad \alpha = e, g . \tag{10.8}$$

When we multiply Eq. (2.6) on the left by  $P_\alpha$ , take the trace, use

$$\text{Tr } P_\alpha (L_a \sigma) = 0 , \quad \alpha = e, g , \tag{10.9}$$

and work out the result for  $\text{Tr } P_\alpha (\Gamma\sigma)$ , as it follows from Eq. (10.5), then we obtain

$$\frac{dn_e}{dt} = -A_e n_e + A_g n_g , \tag{10.10}$$

$$\frac{dn_g}{dt} = -A_g n_g + A_e n_e \quad (10.11)$$

First we notice that the right-hand sides of these equations differ only by a minus sign, and therefore  $n_e + n_g$  is independent of time. The normalization is

$$n_e(t) + n_g(t) = 1 \quad (10.12)$$

which is nothing but  $\text{Tr}\rho(t) = 1$ . Equations (10.10) and (10.11) relate populations only, and they have the form of a set of loss/gain equations. The physical interpretation of these rate equations is that  $A_e n_e$  equals the number of  $|e\rangle \rightarrow |g\rangle$  transitions per unit of time, and  $A_g n_g$  is the number of  $|g\rangle \rightarrow |e\rangle$  transitions per unit of time. Therefore,  $A_e$  and  $A_g$  are the rate constants for  $|e\rangle \rightarrow |g\rangle$  and  $|g\rangle \rightarrow |e\rangle$  transitions, respectively, and they describe spontaneous transitions by this atom in the vacuum near a PC. In the limit  $|P|^2 \rightarrow 0$  the value of  $A_e$  reduces to  $A$ , whereas  $A_g$  vanishes. Then we have ordinary spontaneous decay, with  $A$  as the rate constant. Due to the presence of the PC, the value of  $A$  increases to  $A_e$ , which shows that the PC has the effect of enhancing the spontaneous-decay rate. More interesting is that  $A_g$  acquires a finite value, which implies that there are spontaneous excitations of the atom in vacuum, at a rate  $A_g n_g$ . It is also worth noting that the increase of both  $A_e$  and  $A_g$ , as compared to their empty-space values, equals  $\frac{1}{2}A|P|^2$ . This situation is reminiscent of the coupling of a small system to a nonzero-temperature reservoir, where the increase of the relaxation constants due to the finite temperature is also equal for decay and excitation. An obvious interpretation of Eqs. (10.6) and (10.7) is that the atom will decay

spontaneously with a rate constant equal to  $A$ , but that in addition we have stimulated  $|e\rangle \rightarrow |g\rangle$  and  $|g\rangle \rightarrow |e\rangle$  transitions, which are induced by the PC, and which increase the rate constants for both down and up transitions by the same amount.

#### XI. COMPARISON WITH FLUORESCENT EMISSION

As mentioned in the Introduction, spontaneous decay is intimately related to spontaneous emission because of energy conservation. For the present situation, however, this relation is not too obvious as follows from the fact that there is the possibility of spontaneous excitation. A  $|g\rangle \rightarrow |e\rangle$  transition requires the supply of an amount of energy equal to  $\hbar\omega_0$ , and the process is of course the absorption of a photon. Only the PC, with its two pump beams, can provide this energy.

Elsewhere [19] we have derived an expression for the emitted fluorescent power  $dW/dt$  by an independent method (solving Maxwell's equations without an explicit quantization of the radiation field). We only considered the emission into the positive  $z$ -direction, since only this field can be measured directly by a photomultiplier in the far zone. For the number of emitted photons per unit of time into the  $z > 0$  direction we found

$$\frac{1}{\hbar\omega_0} \frac{dW}{dt} = \frac{1}{2} A n_e + A_g n_g, \quad (11.1)$$

with  $A_g = \frac{1}{2} A |P|^2$ . That the situation is more complicated than for ordinary fluorescence is illustrated with the four diagrams of Fig. 2. The spontaneous contribution  $A$  to  $A_g$ , Eq. (10.6), corresponds to ordinary spontaneous decay, accompanied by the emission of a fluorescent photon. This process happens at



a rate  $An_e$ , and every transition  $|e\rangle \rightarrow |g\rangle$  produces a photon. However, as shown in diagrams (1) and (2), only half the number of photons travels towards the detector in  $z > 0$ , and this gives the term  $\frac{1}{2}An_e$  on the right-hand side of Eq. (11.1). Then, the stimulated contribution to the relaxation constants equals  $\frac{1}{2}A|P|^2$  for both up and down transitions. Diagram (3) of Fig. 2 illustrates the stimulated  $|e\rangle \rightarrow |g\rangle$  transition and the corresponding photon emission. The rate of this process is proportional to  $n_e$ , but the  $n_e$ -term in Eq. (11.1) is already accounted for. Since energy conservation requires the emission of a photon, we conclude that this photon must have frequency  $\bar{\omega}$ , and is therefore indistinguishable from the  $\bar{\omega}$  photons already present in the vacuum. This situation is analogous to atomic decay in a finite-temperature vacuum, where the emitted photons in a stimulated transition are indistinguishable from the black-body radiation. The stimulated  $|g\rangle \rightarrow |e\rangle$  excitations are shown in diagram (4), and they correspond to the term  $A_g n_g = \frac{1}{2}A|P|^2 n_g$  on the right-hand side of Eq. (11.1). Since this term is proportional to  $n_g$ , the atom must be originally in its ground state. An absorption of a vacuum photon with frequency  $\bar{\omega}$  brings the atom to its excited state, and a subsequent decay produces a fluorescence photon. It follows from the rate equations (10.10) and (10.11) that the atom must be in state  $|e\rangle$  after completion of the process, and therefore a second photon with frequency  $\bar{\omega}$  will be absorbed. This three-photon process gives a fluorescent photon with frequency  $2\bar{\omega} - \omega_0$ , whereas the spontaneous-decay process from diagram (1) produces a photon with frequency  $\omega_0$ . With frequency selective detection it should be possible to discriminate between the two processes in an experimental observation of the fluorescence.

## XII. RELAXATION

The set of equations (10.10), (10.11) is easily solved. For a given population of the excited state at  $t = 0$ , the solution for  $t > 0$  is

$$n_e(t) = \frac{A_g}{A_e + A_g} + e^{-(A_e + A_g)t} \left( n_e(0) - \frac{A_g}{A_e + A_g} \right), \quad (12.1)$$

and the population of the ground state follows from Eq. (10.12). For  $t \gg 0$  we obtain the steady-state value

$$n_e(\infty) = \frac{A_g}{A_e + A_g} = \frac{1}{2} \frac{|P|^2}{1 + |P|^2}. \quad (12.2)$$

Without a PC the long-time limit is always  $n_e(\infty) = 0$ , but due to stimulated excitations from the ground state there is a finite probability to find the atom in its excited state. Notice that this probability is determined by the phase-conjugate reflectivity  $|P|^2$  only, and that it is independent of  $A$  (and thereby independent of the dipole moment  $\mu_{eg}$ ). It can be shown [37], however, that this is an artefact of a two-state model. Also notice that the time constant of the relaxation is the inverse of

$$A_e + A_g = A(1 + |P|^2), \quad (12.3)$$

which is larger than  $A$ . Consequently, an atom near a PC relaxes faster than in empty space.

In the steady state the photon emission rate from Eq. (11.1) becomes

$$\frac{1}{\hbar\omega_0} \frac{dW}{dt} (\tau \rightarrow \infty) = \frac{1}{2} A |P|^2 \frac{3 + |P|^2}{1 + |P|^2}, \quad (12.4)$$

which is nonzero for  $|P|^2 \neq 0$ . An atom near a PC continuously emits radiation, both due to ordinary decay (diagram (1) from Fig. 2) and due to three-photon processes (diagram (4)). From Eq. (12.2) we derive

$$A_g n_g(\infty) = (2 + |P|^2) \left( \frac{1}{2} A n_e(\infty) \right), \quad (12.5)$$

which shows that the three-photon processes produce a factor of  $2 + |P|^2$  more photons in  $z > 0$  than ordinary decay from the excited state.

The equation of motion (2.6) is also easily solved for the time evolution of the coherences. It appears that the equation for  $\langle e | \rho(t) | g \rangle$  does not couple to the rate equations for the populations, and the general solution is found to be

$$\langle e | \rho(t) | g \rangle = e^{-\frac{1}{2}(A_e + A_g)t} e^{-i\omega_0 t} \langle e | \rho(0) | g \rangle, \quad (12.6)$$

for a given initial state  $\rho(0)$ . The relaxation time for the coherences is twice as long as the relaxation time for the populations, as could be expected from the analogy with thermal relaxation.

### XIII. DEGENERATE TWO-LEVEL ATOM AND TRANSPARENT PC

Although in certain experiments one can prepare an atom so as to behave effectively as a two-state atom [38], in general this is an oversimplification

of the situation. In particular, for an atom near a PC we have the four processes of Fig. 2 between any set of two states with a nonvanishing transition dipole moment  $\mu_{eg}$ . When the excited level is degenerate, for instance, then there are stimulated excitations (diagram (4)) from a ground state to any of the excited states. Also, the assumption of an ideal PC is not very realistic, although it is adequate to study the fundamental processes from Fig. 2. In this section we consider the realistic situation of a transparent PC, as discussed in Sec. VIII.D, and we assume that the pump frequency is reasonably close to an atomic resonance  $\omega_0$ , so that  $y_{\perp}(\omega)$  and  $y_{\parallel}(\omega)$  from Eqs. (8.14) and (8.15), respectively, are nonzero for  $\omega = \omega_0$ . Then the vacuum correlation function at  $\omega = \omega_0$  and  $\omega = -\omega_0$  is

$$\tilde{F}_{\alpha}(\omega_0) = \eta(\omega_0)(1 + y_{\alpha}(\omega_0)) \quad , \quad (13.1)$$

$$\tilde{F}_{\alpha}(-\omega_0) = \eta(\omega_0)y_{\alpha}(\omega_0) \quad , \quad (13.2)$$

with  $\alpha = \perp$  or  $\parallel$ . Here it is also assumed that  $|P_{\sigma}|^2$  is symmetric around  $\bar{\omega}$  in its frequency dependence. From now on we write  $\eta$ ,  $y_{\perp}$  and  $y_{\parallel}$  since the frequency  $\omega_0$  is fixed.

With  $j_e$  and  $j_g$  the angular momentum quantum numbers of the excited level and ground level, respectively, we can write  $|j_e m_e\rangle$  and  $|j_g m_g\rangle$  for their magnetic substates. As the quantization axis for the definition of the degeneracies we take the normal to the surface ( $z$ -axis). The excited level and ground level are  $(2j_e + 1)$ - and  $(2j_g + 1)$ -fold degenerate, and since the dipole moment matrix element vanishes for  $j_e - j_g = 0$ , the minimum number of atomic states is four (for  $j_e - j_g = \frac{1}{2}$  or  $j_e = 1, j_g = 0$  or  $j_e = 0, j_g = 1$ ).

The generalization of the atomic raising operator from Eq. (10.3) for a degenerate two-level atom is defined as

$$d_r = \sum_{m_e m_g} (j_{g m_g} 1 r | j_{e m_e}) | j_{e m_e} \rangle \langle j_{g m_g} | , \quad r = -1, 0, 1 , \quad (13.3)$$

in terms of which  $\underline{\mu}^{(-)}$  can be expressed as

$$\underline{\mu}^{(-)} = \frac{\langle j_e \| \underline{\mu} \| j_g \rangle}{\sqrt{2j_e + 1}} \sum_r d_r \underline{e}_r^* . \quad (13.4)$$

Here,  $\underline{e}_r$  is a spherical unit vector with respect to the z-axis, and  $\underline{\mu}^{(+)}$  equals the Hermitian conjugate of the right-hand side.

The spontaneous-decay operator  $\Gamma$  is expressed in the Cartesian components of  $\underline{\mu}^{(+)}$  and  $\underline{\mu}^{(-)}$  in Eq. (9.13), and with Eq. (13.4) and its Hermitian conjugate we can simplify the result considerably. In analogy to Eq. (10.5) for a two-state atom, we now obtain

$$\begin{aligned} \Gamma \sigma = & \frac{1}{2} \sum_r A_r^e (d_r d_r^\dagger \sigma + \sigma d_r d_r^\dagger - 2d_r^\dagger \sigma d_r) \\ & + \frac{1}{2} \sum_r A_r^g (d_r^\dagger d_r \sigma + \sigma d_r^\dagger d_r - 2d_r \sigma d_r^\dagger) . \end{aligned} \quad (13.5)$$

The similarity with Eq. (10.5) is even more obvious if we write  $P_e = dd^\dagger$ ,  $P_g = d^\dagger d$  in Eq. (10.5), which holds for a two-state atom (but not for a degenerate two-level atom). In Eq. (13.5) we have introduced the quantities

$$A_r^e = A(1 + y_r) \quad , \quad (13.6)$$

$$A_r^g = Ay_r \quad , \quad (13.7)$$

for  $r = -1, 0, 1$ , where

$$A = \frac{\omega_o^3}{3\pi\epsilon_o\hbar c^3} \frac{|\langle j_e \| \mu \| j_g \rangle|^2}{2j_e + 1} \quad (13.8)$$

is the Einstein coefficient for spontaneous decay of this degenerate two-level atom in empty space. Furthermore, we write

$$y_r = \begin{cases} y_{\perp} & , \quad \text{for } r = 0 \\ y_{\parallel} & , \quad \text{for } r = \pm 1 \end{cases} \quad (13.9)$$

The quantities  $A_r^e$  and  $A_r^g$  are identical in form to the relaxation constants  $A_e$  and  $A_g$  from Eqs. (10.6) and (10.7), respectively, but their interpretation is slightly different, as we shall see later on.

The right-hand side of Eq. (13.5) is probably the most compact representation of the operator  $\Gamma$ . As the first step towards the evaluation of its matrix elements, we substitute the expressions for  $d_r$  and  $d_r^\dagger$ , and use the fact that a Clebsch-Gordan coefficient  $(j_g m_g 1 r | j_e m_e)$  can only be nonzero if  $m_g + r = m_e$ . In terms of the new constants

$$A_{m_e} = \sum_{m_g, r} A_r^e (j_g m_g 1 r | j_e m_e)^2 \quad , \quad (13.10)$$

$$A_{mg} = \sum_{m_e r} A_r^g (j_{g m_g} 1 r | j_{e m_e})^2, \quad (13.11)$$

the operator  $\Gamma$  attains the form

$$\begin{aligned} \Gamma \sigma = & \frac{1}{2} \sum_{m_e} A_{m_e} (|j_{e m_e} \times j_{e m_e} | \sigma + \sigma | j_{e m_e} \times j_{e m_e} |) \\ & + \frac{1}{2} \sum_{m_g} A_{m_g} (|j_{g m_g} \times j_{g m_g} | \sigma + \sigma | j_{g m_g} \times j_{g m_g} |) \\ & - \sum_{\substack{m_e m_g r \\ m'_e m'_g}} (j_{g m_g} 1 r | j_{e m_e}) (j_{g m'_g} 1 r | j_{e m'_e}) \\ & \times (A_r^e \langle j_{e m_e} | \sigma | j_{e m'_e} \rangle | j_{g m_g} \times j_{g m'_g} | \\ & + A_r^g \langle j_{g m_g} | \sigma | j_{g m'_g} \rangle | j_{e m_e} \times j_{e m'_e} |) , \end{aligned} \quad (13.12)$$

which allows a direct evaluation of matrix elements of  $\Gamma \sigma$  in terms of matrix elements of  $\sigma$ .

#### XIV. EQUATION OF MOTION

With Eq. (13.12) we can expand the equation of motion (2.6) for  $\rho(t)$  in matrix elements. This gives the linear set of coupled differential equations

$$\begin{aligned} \frac{d}{dt} \langle j_{e m_e} | \rho | j_{e m'_e} \rangle = -\frac{1}{2} (A_{m_e} + A_{m'_e}) \langle j_{e m_e} | \rho | j_{e m'_e} \rangle \\ + \sum_{m_g m'_g r} A_r^g (j_{g m_g} 1 r | j_{e m_e}) (j_{g m'_g} 1 r | j_{e m'_e}) \langle j_{g m_g} | \rho | j_{g m'_g} \rangle, \end{aligned} \quad (14.1)$$

$$\begin{aligned} \frac{d}{dt} \langle j_{g m_g} | \rho | j_{g m'_g} \rangle = -\frac{1}{2} (A_{m_g} + A_{m'_g}) \langle j_{g m_g} | \rho | j_{g m'_g} \rangle \\ + \sum_{m_e m'_e r} A_r^e (j_{g m_g} 1 r | j_{e m_e}) (j_{g m'_g} 1 r | j_{e m'_e}) \langle j_{e m_e} | \rho | j_{e m'_e} \rangle, \end{aligned} \quad (14.2)$$

$$\frac{d}{dt} \langle j_{e m_e} | \rho | j_{g m_g} \rangle = -\left\{ \frac{1}{2} (A_{m_e} + A_{m_g}) + i\omega_0 \right\} \langle j_{e m_e} | \rho | j_{g m_g} \rangle, \quad (14.3)$$

$$\frac{d}{dt} \langle j_{g m_g} | \rho | j_{e m_e} \rangle = -\left\{ \frac{1}{2} (A_{m_e} + A_{m_g}) - i\omega_0 \right\} \langle j_{g m_g} | \rho | j_{e m_e} \rangle. \quad (14.4)$$

As a first observation, we notice that Eqs. (14.3) and (14.4) for the coherences between an excited state and a ground state do not couple with the rest of the set. The solution of Eq. (14.3) is identical to the solution (12.6) for a two-state atom, provided that we replace  $A_e + A_g$  by  $A_{m_e} + A_{m_g}$ . For the decay of the coherence between two magnetic substates of the excited level, we have Eq. (14.1) with  $m_e \neq m'_e$ , and it follows that the time evolution of  $\langle j_{e m_e} | \rho | j_{e m'_e} \rangle$  is influenced by the decay of the coherences  $\langle j_{g m_g} | \rho | j_{g m'_g} \rangle$  between the various substates of the ground level. Note that the condition



$m_e \neq m'_e$  makes the term with  $m_g = m'_g$  under the summation sign equal to zero, and therefore also the time evolution of these coherences is independent of the time evolution of the populations. The same conclusions hold for the coherences in the ground level, as described Eq. (14.2) with  $m_g \neq m'_g$ .

When we set  $m'_e = m_e$  in Eq. (14.1) and  $m'_g = m_g$  in Eq. (14.2), and then perform the summations over  $m'_g$  and  $m'_e$ , respectively, we get

$$\begin{aligned} \frac{d}{dt} \langle j_{e m_e} | \rho | j_{e m_e} \rangle &= -A_{m_e} \langle j_{e m_e} | \rho | j_{e m_e} \rangle \\ &+ \sum_{m_g r} A_r^g (j_{g m_g} 1 r | j_{e m_e})^2 \langle j_{g m_g} | \rho | j_{g m_g} \rangle, \end{aligned} \quad (14.5)$$

$$\begin{aligned} \frac{d}{dt} \langle j_{g m_g} | \rho | j_{g m_g} \rangle &= -A_{m_g} \langle j_{g m_g} | \rho | j_{g m_g} \rangle \\ &+ \sum_{m_e r} A_r^e (j_{g m_g} 1 r | j_{e m_e})^2 \langle j_{e m_e} | \rho | j_{e m_e} \rangle. \end{aligned} \quad (14.6)$$

This set only contains the populations of the various substates, and it has the form of a set of rate equations. The first term on the right-hand side of Eq. (14.5) accounts for the loss of population of  $|j_{e m_e}\rangle$ , due to transitions to the various ground states, and every  $m_g$ -term in the summation gives the gain of population of  $|j_{e m_e}\rangle$ , due to stimulated excitations to this level and from this particular ground state. This is in complete analogy to Eq. (10.10) for a two-state atom. The same interpretation holds for Eq. (14.6), but with the role of  $|j_{e m_e}\rangle$  and  $|j_{g m_g}\rangle$  reversed. Now that we have identified the structure of Eqs. (14.5) and (14.6), we can interpret the significance of the relaxation parameters  $A_{m_e}$ ,  $A_{m_g}$ ,  $A_r^e$  and  $A_r^g$ . From Eq. (14.6) it follows that

$\sum_r A_r^e (j_{g g}^{m_g} | r | j_{e e}^{m_e})^2$  is the rate constant for the transition  $|j_{e e}^{m_e}\rangle \rightarrow |j_{g g}^{m_g}\rangle$ , and the transition rate equals this constant times the population of  $|j_{e e}^{m_e}\rangle$ . The appearance of the Clebsch-Gordan coefficient signifies the dipole selection rule. When we sum this rate constant over  $m_g$  we get  $A_{m_e}$ , as defined by Eq. (13.10), and from Eq. (14.5) we see that  $A_{m_e}$  must equal the rate constant for the loss of population of  $|j_{e e}^{m_e}\rangle$ , and independent of which ground state this population ends up in. This makes the interpretation of both rate constants consistent. Similarly,  $\sum_r A_r^g (j_{g g}^{m_g} | r | j_{e e}^{m_e})^2$  is the rate constant for the  $|j_{g g}^{m_g}\rangle \rightarrow |j_{e e}^{m_e}\rangle$  transition, and summed over all values  $m_e$  of the final state this gives the rate constant  $A_{m_g}$  for the loss of population of  $|j_{g g}^{m_g}\rangle$ .

It is interesting to sum Eqs. (14.5) and (14.6) over  $m_e$  and  $m_g$ , respectively. This yields

$$\frac{dn_e}{dt} = - \sum_{m_e} A_{m_e} \langle j_{e e}^{m_e} | \rho | j_{e e}^{m_e} \rangle + \sum_{m_g} A_{m_g} \langle j_{g g}^{m_g} | \rho | j_{g g}^{m_g} \rangle, \quad (14.7)$$

$$\frac{dn_g}{dt} = - \sum_{m_g} A_{m_g} \langle j_{g g}^{m_g} | \rho | j_{g g}^{m_g} \rangle + \sum_{m_e} A_{m_e} \langle j_{e e}^{m_e} | \rho | j_{e e}^{m_e} \rangle, \quad (14.8)$$

where

$$n_\alpha = \sum_{m_\alpha} \langle j_{\alpha \alpha}^{m_\alpha} | \rho | j_{\alpha \alpha}^{m_\alpha} \rangle, \quad \alpha = e, g \quad (14.9)$$

are the populations of the two levels, independent of how this population is

distributed over the various substates. Only the relaxation constants  $A_{m_e}$  and  $A_{m_g}$  appear in Eqs. (14.7) and (14.8), as is consistent with our earlier interpretation of these parameters. It also follows immediately that  $n_e(t) + n_g(t)$  is independent of time, as it should be.

## XV. MISCELLANEA

### A. Sum rules and symmetry

With the well-known properties of Clebsch-Gordan coefficients, we derive from Eq. (13.10)

$$\frac{1}{2j_e + 1} \sum_{m_e} A_{m_e} = \frac{1}{3} \sum_r A_r^e = \frac{1}{3} A_{\perp}^e + \frac{2}{3} A_{\parallel}^e, \quad (15.1)$$

where the  $\perp$  and  $\parallel$  notation is related to  $r$ -values as in Eq. (13.9). Since there are  $2j_e + 1$  values of  $m_e$  and three values of  $r$ , the first equality states that the average value of  $A_{m_e}$  equals the average value of  $A_r^e$ . Then the second equality expresses that there are twice as much parallel degrees of freedom ( $x$  and  $y$ ) for the orientation of the dipole moment than there are for the perpendicular direction (only  $z$ ). When we sum  $A_{m_g}$  over  $m_g$  we find

$$\frac{1}{2j_e + 1} \sum_{m_g} A_{m_g} = \frac{1}{3} \sum_r A_r^g = \frac{1}{3} A_{\perp}^g + \frac{2}{3} A_{\parallel}^g. \quad (15.2)$$

Here, the left-hand side is not the average value of  $A_{m_g}$  when  $j_e \neq j_g$ . We shall show in Sec. XV.C why the factor in front of the summation sign in Eq. (15.2) must be  $1/(2j_e + 1)$ , and not  $1/(2j_g + 1)$ .

Another interesting relation, which follows directly from the properties of Clebsch-Gordan coefficients, in combination with Eqs. (13.10) and (13.11), is

$$A_{-m_e} = A_{m_e}, \quad A_{-m_g} = A_{m_g}. \quad (15.3)$$

It can be shown [31], that the sum rules (15.1) and (15.2) and the symmetry relations (15.3) are entirely geometrical. They can be derived from the symmetry of the vacuum field for rotations about the z-axis and reflections in a plane through that axis. Since these symmetries hold quite generally near a flat surface, these results are independent of the details of the four-wave mixing process.

#### B. Fluorescence

For a degenerate two-level atom, the number of emitted photons per unit of time in the  $z > 0$  direction equals [19]

$$\frac{1}{\hbar\omega_0} \frac{dW}{dt} = \frac{1}{2} A \text{Tr} \rho(t) \sum_r (d_r d_r^\dagger + 2y_r d_r^\dagger d_r) \quad (15.4)$$

When we substitute the expressions for  $d_r$  and  $d_r^\dagger$ , evaluate the trace, and use the definition of  $A_{m_g}$ , then Eq. (15.4) reduces to

$$\frac{1}{\hbar\omega_0} \frac{dW}{dt} = \frac{1}{2} A n_e + \sum_{m_g} A_{m_g} \langle j_{g m_g} | \rho | j_{g m_g} \rangle \quad (15.5)$$

where  $n_e$  is the total population of the excited level. We find again that the first contribution,  $4An_e$ , is ordinary fluorescence, which is emitted independent of the presence of the PC. Every term under the summation sign corresponds to the occurrence of a three-photon process between the particular state  $|j_{m_g}\rangle$  and the excited level. It appears that the contributions from the various excited states combine in the single rate constant  $A_{m_g}$ , and therefore the rate of emission of fluorescent photons due to a population of  $|j_{m_g}\rangle$  is equal to the rate of spontaneous transitions from  $|j_{m_g}\rangle$  to the excited level, as could be expected.

### C. Ideal PC

For the case of a degenerate two-level atom and a transparent PC, we can find the limit of an ideal PC by setting

$$y_{\perp} = y_{\parallel} = 4|P|^2. \quad (15.6)$$

With Eqs. (13.6) and (13.7) we then obtain  $A_r^e$  and  $A_r^g$ , which become independent of  $r$ . In Eqs. (13.10) and (13.11) we can then perform the summations, and it follows that  $A_{m_e}$  and  $A_{m_g}$  become independent of  $m_e$  and  $m_g$ , respectively. Hence we define  $A_e$  and  $A_g$  as

$$A_e = A_{m_e}, \quad A_g = A_{m_g}, \quad (15.7)$$

and they are found to be

$$A_e = A_r^e = A(1 + \frac{1}{2}|P|^2) \quad , \quad (15.8)$$

$$A_g = \frac{2j_e + 1}{2j_g + 1} A_r^g = \frac{1}{2} A|P|^2 \frac{2j_e + 1}{2j_g + 1} \quad . \quad (15.9)$$

Equation (13.5) reduces to

$$\begin{aligned} \Gamma\sigma = & \frac{1}{2} A_e (P_e \sigma + \sigma P_e - 2 \sum_r d_r^\dagger \sigma d_r) \\ & + \frac{1}{2} A_g (P_g \sigma + \sigma P_g - 2 \frac{2j_g + 1}{2j_e + 1} \sum_r d_r \sigma d_r^\dagger) \quad , \end{aligned} \quad (15.10)$$

in terms of the projectors

$$P_\alpha = \sum_{m_\alpha} |j_{\alpha m_\alpha} \rangle \langle j_{\alpha m_\alpha}| \quad , \quad \alpha = e, g \quad . \quad (15.11)$$

For the populations we now find

$$\frac{dn_e}{dt} = -A_e n_e + A_g n_g \quad , \quad (15.12)$$

$$\frac{dn_g}{dt} = -A_g n_g + A_e n_e \quad , \quad (15.13)$$

which is identical in form to Eqs. (10.10) and (10.11). This shows that  $A_e$  and  $A_g$  are the rate constants for the depopulation of the excited level and the ground level, respectively, irrespective of how this population is distributed over the various states. Notice that the excited level and the

ground level have the same relaxation constant as their individual states  $|j_e m_e\rangle$  and  $|j_g m_g\rangle$ , as follows from Eq. (15.6). It can also be shown that the coherences between the two levels decay with  $\frac{1}{2}(A_e + A_g)$  as inverse relaxation constant, and that the fluorescence rate reduces to

$$\frac{1}{\hbar\omega_0} \frac{dW}{dt} = \frac{1}{2} A_e n_e + A_g n_g \quad (15.14)$$

We conclude that in the limit of an ideal PC this degenerate two-level atom behaves similarly as a two-state atom, at least as far as the time evolution of the atomic density operator is concerned, or the fluorescent yield. Notice that also the expression (10.6) and (15.8) for the relaxation constant of the excited state are identical in form, but that  $A_g$  in Eq. (15.9) acquires an additional factor  $(2j_e + 1)/(2j_g + 1)$ , as compared to  $A_g$  in Eq. (10.7). This factor equals the ratio of the number of excited states and the number of ground states, and it also appears explicitly in expression (15.10) for  $\Gamma$  and in the sum rule (15.2) for the average of  $A_{mg}$ . It takes into account the difference between the number of excited states and the number of ground states, and it leads to the correct form of the set of rate equations (15.12), (15.13).

## XVI. CONCLUSIONS

We have studied the relaxation of an atom in the vicinity of a PC. It appeared that the atomic relaxation operator  $\Gamma$  could be expressed in the two vacuum correlation functions  $\tilde{f}_\perp(\omega)$  and  $\tilde{f}_\parallel(\omega)$ , which contain all necessary information about the medium and the four-wave mixing process. We were able to express these functions in terms of the Fresnel reflection and transmission

coefficients for plane waves, and the normal distance between the atom and the surface of the medium. These coefficients depend in a complicated way on both the linear and nonlinear interactions, the polarization and the angle of incidence. For instance, it was shown in a recent experiment [39] that even the Fresnel coefficient for the reflection of the specular wave is strongly affected by the nonlinear interaction. We found that the correlation functions could be expressed in terms of the parameter functions  $x_\alpha$ ,  $y_\alpha$  and  $z_\alpha$ , with  $\alpha = \perp$  or  $\parallel$ .

Subsequently we worked out the model case of a two-state atom, in combination with the approximation of an ideal PC. The relaxation operator  $\Gamma$  then assumes the simple form (10.5). We obtained the relaxation constants  $A_e$  and  $A_g$  for the excited state and the ground state, respectively, and we identified the four processes which are responsible for the relaxation (Fig. 2). Ordinary spontaneous fluorescent emission in combination with an  $|e\rangle \rightarrow |g\rangle$  transition gives the usual contribution to  $A_e$  (equal to  $A$ ). Furthermore, stimulated  $|e\rangle \rightarrow |g\rangle$  transitions in combination with the emission of a photon, and stimulated three-photon processes which excite the atom contribute to the relaxation. The rate constants for both (down and up) stimulated processes are identical, at least in the limit of an ideal PC in combination with a two-state atom. This could be expected from the analogy with stimulated thermal transitions, although the physical mechanism is different. These stimulated processes always increase the values of the relaxation constants, thereby decreasing the relaxation time of the system. The three-photon processes are responsible for a finite population of the excited state in the limit  $t \rightarrow \infty$ . We found that both  $A_e$  and  $A_g$  are independent of the atom-surface distance, which resembles similar predictions by Agarwal [40] and Cook and Milonni [41].



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# FIGURE CAPTIONS

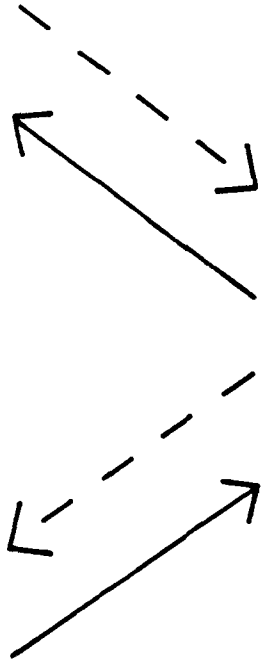
Fig. 1. A plane wave is incident upon a layer of nonlinear material with thickness  $\Delta$ . At the boundary  $z = 0$  this wave partially reflects into the specular direction (r-wave), and partially travels into the medium. At  $z = -\Delta$  there is again partial reflection and partial transmission, which gives rise to multiple reflections inside the medium and the t-wave in  $z < -\Delta$ . These waves are indicated by the solid arrows, where the arrow head indicates the propagation direction. These waves are already present for a linear medium. Due to the nonlinear interaction, the two waves inside the medium couple each with a counterpropagating negative-frequency wave (provided that the incident field has a positive frequency), which leave the medium at the boundaries  $z = 0$  and  $z = -\Delta$  as the pc-wave and the nl-wave, respectively. These negative-frequency waves are shown as broken arrows. For a dielectric constant equal to unity, the r-wave and the nl-wave, including the two corresponding waves in the medium, disappear to a good approximation. The figure illustrates the situation for which the incident wave comes from the positive  $z$ -direction. As explained in the text, there are also vacuum waves incident from the region  $z < -\Delta$ , and they give rise to nl-waves and t-waves in  $z > 0$ .

Fig. 2. Illustration of the four processes which contribute to the atomic relaxation and to the emission of observable fluorescence. The rates for the processes are indicated, and the wiggly arrows show pictorially the propagation direction of the photons involved. Diagram (1) represents ordinary spontaneous decay, together with an  $|e\rangle \rightarrow |g\rangle$  transition of the atom. The fluorescent photon travels into the direction of the detector and contributes to the fluorescent yield, whereas the atomic decay gives rise to relaxation. The same process is shown in diagram (2), except that the photon here travels towards the PC. Therefore, this process contributes to the

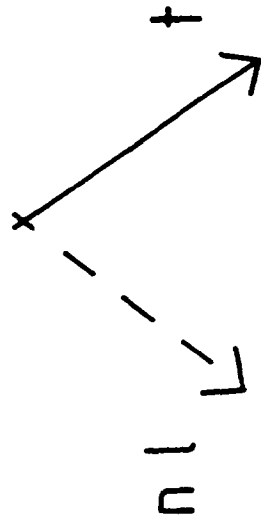
relaxation, but not to the observable fluorescence. Diagram (3) looks the same as diagram (1), except that the rate is multiplied by  $|P|^2$ . We identify this process as stimulated decay combined with the emission of a photon. This photon also travels into the positive z-direction, because it is a t-wave, but it does not contribute to the fluorescence. In the three-photon process from diagram (4) the atom makes the transitions  $|g\rangle \rightarrow |e\rangle \rightarrow |g\rangle \rightarrow |e\rangle$ , where the first and the last transitions are accompanied by the absorption of a photon with frequency  $\bar{\omega}$ . During the decay in between these absorptions, the atom emits a photon with frequency  $2\bar{\omega} - \omega_0$  in the positive z-direction. Notice that emitted photons in a stimulated transition always travel in the positive z-direction.



$z = 0$



$z = -\Delta$



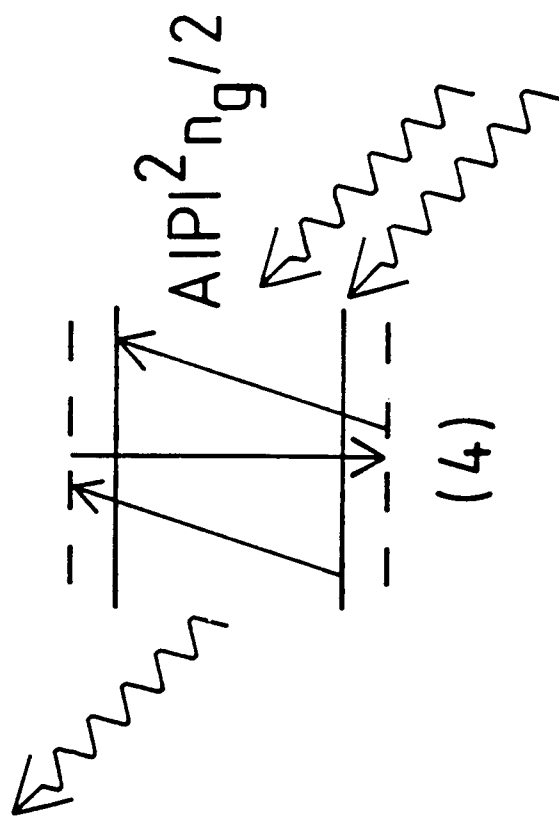
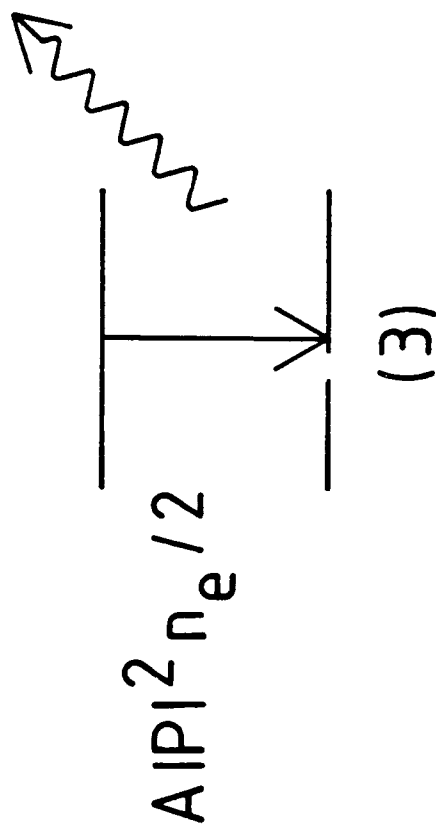
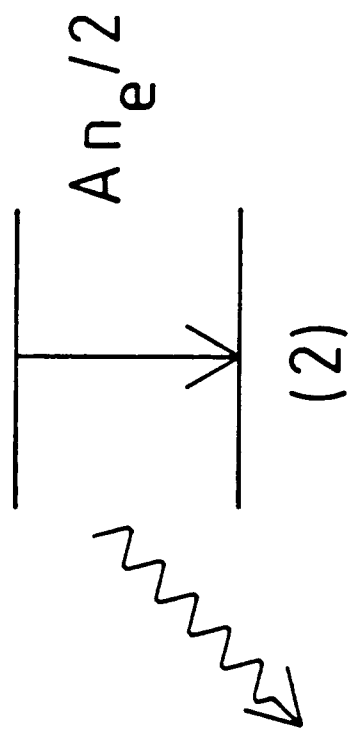
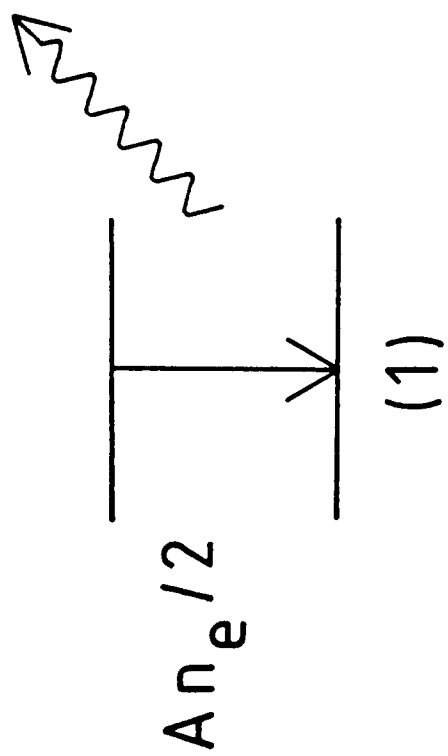


Fig.2 HFA/TFG